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Assessing heavy metal pollution in paddy soil from coal mining area, Anhui, China

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Abstract Heavy metal pollution in agricultural soil has negative impact on crop quality and eventually on human health. A total of 24 top soil samples were collected from paddy field near the Zhangji Coal Mine in Huainan City, Anhui Province. Seven heavy metals (Cu, Zn, As, Cr, Cd, Pb, and Ni) were selected to evaluate the pollution status through total content and chemical speciation, geo-accumulation index (I_{geo}), and risk assessment code (RAC) and investigate leaching behavior of heavy metals under simulated rainfall. The results of present study indicated that mining activities were responsible for elevated Cu and Cd in surrounding paddy soil. Based on the results of chemical speciation, most heavy metals were associated with the residual fraction, and the environmental risk of heavy metals in soil was sequenced as Pb > Cd > Ni > As > Zn > Cu > Cr. It revealed that Pb in soil would pose a higher environmental risk due to its higher reducible fraction, then followed by Cd, Ni, As, and Zn, which would pose a medium risk. The result of simulated rainfall leaching analysis showed that heavy metals could be categorized into two groups: concentrations of Cu, Ni, Cd, Zn, and Cr in the leachates displayed a continuous decrease tendency with the increase in accumulative simulated rain volume; whereas leachable tendency of As and Pb was enhanced with increasing leaching time and rain volume. Generally, the leaching percentage of heavy metals followed the sequence of As > Zn > Ni > Cd > Cr > Cu > Pb. More attention should be paid to the higher environmental risk of Pb and higher leaching percentage of As with regard to ecosystem safety and human health.

Keywords Agricultural soil · Heavy metal · Environmental assessment

Introduction

The global demand for energy was increased by 2.1% in 2017, among which 40% of the growth was generated from China and India (Tollefson, 2017; IEA. 2018). China was the largest coal producer and consumer throughout the world, where more than half of the fuel and energy source was coal owing to its high abundance and low cost (You and Xu 2010; Mi et al. 2018; Yan et al. 2018). However, the annual cost for subsequent restoration and remediation after coal exploitation and utilization was estimated to be 1400 billion RMB in China (Epstein et al, 2011; Hu et al. 2014), which posed

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a financial burden to the government and society. Furthermore, environmental problems associated with coal utilization, such as heavy metal pollution, particulate matter ($PM_{2.5}$), polycyclic aromatic hydrocarbons (PAHs) emission, and acid rain, result in detrimental environmental effects, which would further threat human health, either directly or indirectly (Tian et al. 2010; Li et al. 2014a, b; Pandey et al. 2016; Sun et al. 2018; Landrigan et al. 2018a, b; Ouyang et al. 2018).

Heavy metals were generally referred to elements with high density (> 5 g/cm^3), such as chromium, mercury, zinc, lead, cadmium, copper, and nickel, which was toxic, non-biodegradable and could be biomagnificated through food chains (Jing et al. 2018). Presence of environmental pollution event such as the itai-itai disease in Japan and Cd-contaminated rice as well as arsenicosis in China indicated that human health was chronically impacted by heavy metals in environment (Lu et al. 2015). It was demonstrated by the Ministry of Environmental Protection of China in 2011 that coal mining was the predominant source of heavy metals (such as Pb, Hg, Cr, Cd, and As) exposure in China. According to Finkelman (Finkelman 1999), coal contained more than 80 trace elements including various heavy metals, which could be released into the surrounding environment during mining processes. Therein, soil contaminated with heavy metals in mining area might have negative effect on microbial activity and nutrients availability (Chibuike and Obiora 2014; Zhang et al. 2015). In addition, bioavailable heavy metals in agricultural soil could be accumulated by crops, and then transferred into human body through daily food consumption. Therefore, monitoring and assessment of heavy metal pollution in agricultural soil in accompany with soil fertility test for soil quality assessment in coal mining area was practically demanded (Wang et al. 2018).

Generally, soil was a sink for heavy metals, where a range of processes including precipitation, adsorption, and redox reaction could lower the mobility of heavy metals. However, once the levels of heavy metals exceeded the soil environmental capacity or the soil environment changed (such as pH and redox potential), heavy metals could be released and available for organisms. Thus, soil was also an important source of heavy metals. Soil played an important role in human food supply. It can satisfy approximately 90% of the human calorie demand directly and indirectly (Bünemann et al. 2018). However, approximately 17% of the arable land on earth had degraded due to deforestation and excessive farming practices, which had resulted in a 0.3% diminished output of global crop production (Kaiser 2004; Reich and Eswaran 2004). According to the report of the latest national soil pollution survey in 2014, more than 19% of the farmland soil exceeded the environmental quality standards for soil, especially being highly contaminated with cadmium, mercury, and arsenic. Consequently, a portion of heavy metals in agricultural soil could be bioaccumulated in crops and finally have chronic and deleterious impact on human health through food chains (Landrigan et al. 2018a, b; Jing et al. 2018; Zhang et al. 2019; Cheng et al. 2017). It was estimated that more than 12 billion tons of crops had been contaminated with heavy metals every year in China, leading to an annual financial loss of 20 billion RMB (Luo and Teng 2006; Guan et al. 2017). Therefore, soil can definitely have adverse effect on economic development and human health with regard to its exposure to heavy metals from various sources (Brevik and Burgess 2014; Brevik et al. 2018).

Nowadays, studies investigating heavy metals in soil were mainly focused on the total content, distribution pattern, and associated environmental and ecological risk assessment. However, as for the soil from the mining area, the source of heavy metals was more complicated, which can result from not only the release during coal exploitation but also the atmospheric deposition originating from coal-fired power plants as well as the dispersion during the process of coal transportation (Luo et al. 2009). Previous studies on coal mining-affected agricultural soil revealed that the levels of heavy metals (such as Mn, Zn, As, and Pb) exceeded corresponding toxic limit standards (Bhuiyan et al. 2010; Teixeira et al. 2001; Zhou et al. 2007). Nevertheless, total content even the chemical speciation characterization of heavy metals in soil was not sufficient for an integrated pollution assessment, especially in coal mining area. Knowledge systematically regarding chemical speciation and leaching behaviors of heavy metals was of great importance in evaluating environmental risk, which also could provide useful information for soil remediation strategies. Therefore, the objective of present study was to investigate the pollution status of heavy metals in agricultural soil from mining area through the total content and chemical speciation analysis. Furthermore, the leaching behaviors of heavy metals under simulated rainfall were also discussed for integrated environmental assessment (Fig. 1).

Materials and methodology

Sample collection

Samples for present study were collected from a site near the Zhangji Coal Mine, Huainan City, Anhui province. Huainan City was located in the central Anhui Province (32° 23.3′–33° 0.5′ N, 116° 21.0′–117° 12.5′ E), of which the coal resource accounted for 19% of the national coal reserve (Sun et al. 2010). Zhangji Coal Mine began production in 2001, with an annual yield of 12.4 million tons of raw coal. Our previous studies indicated that the coal from the Zhangji Coal Mine exhibited high content of As and Cu (Li et al. 2013).

In accordance with the standard procedures for soil sampling, processing, and reposition (NY/T 1121.1-2006), a total of 24 surface soil samples (0–20 cm) were collected from paddy field near the coal mine. All the samples were collected using a stainless-steel spade and stored in clean self-locking polyethylene bags. After returning to the laboratory, samples were air-dried and pulverized to a particle size less than 2 mm in diameter for physico-chemical analyses. Soil pH, cation exchange capacity (CEC), total nitrogen (TN), total phosphorus (TP), and soil organic matter (SOM) were measured according to the risk control standard for soil contamination of agricultural land (GB 15618-2018). The percentages of sand, silt, and clay were determined using the dry sieving method (Sun and Chen 2018).

Sequential extraction

Chemical speciation of heavy metals was determined by a four-step sequential extraction procedure proposed by European Community of Reference (BCR), a detailed description of which was reported previously (Dai et al. 2018; Li et al. 2019). Accordingly, four fractions were partitioned: F1-acid soluble, bound to carbonate and cation exchange site (1.0 g soil sample with 0.11 mol/L HAc shaked for 16 h at 22 ± 5 °C); F2-reducible, bound to Fe-Mn oxides (with 0.5 mol/L NH2OH·HCl shaked for 16 h at 22 ± 5 °C); F3-oxidizable, bound to organic matter and sulfides (with 8.8 mol/L H2O2 shaked for 1 h at 85 ± 2 °C and 1 mol/L NH4Ac shaked for 1 h at 85 ± 2 °C); and F4- remaining residue (with 6 mol/L HCl and 14 mol/L HNO3 left overnight at 20 °C).

Simulated rainfall leaching experiment

Based on the monitoring results for the chemical composition of precipitation in Huainan City, simulated rainfall was prepared with a solution of SO_4^{2-}/NO_3^- in the 5.2:1 ratio, and with a pH of 6.28. The average precipitation was estimated to be 922 mm. The leaching experiment was conducted in PVC cylinders at room temperature, wherein 5 g of quartz sand was placed at the bottom, on top of which, 20 g of air-dried soil sample was added, and then another 5 g quartz sand was placed on top of the soil sample in order to minimize soil migration. Prior to the leaching experiment, deionized water was used to attain the field-holding capacity. Each 35-ml simulated rain was pumped into the cylinder at a rainfall intensity of 0.2 ml/h for every 48 h by ten times. The total amount of leaching solution was 350 mL, which was equivalent to the precipitation in this city over a period of 1 year. Ten leachates were collected for analysis after centrifugation (at 3500 rpm for 20 min) and filtration (through a 0.45-μm cellulose membrane filter).

Heavy metal determination

According to Chinese Ministry of Environmental Protection (CMEP) method HJ 781-2016 and HJ 803-2016 (CMEP. 2016a, b), all the samples were digested and measured using inductively coupled plasma-optical emission spectrometry (ICP-OES). Soil samples, together with the reference standard (GB W07403), were digested using a concentrated acid mixture of HNO3:HCl = 1:3 (in volume). Thereafter, digested solution was diluted and stored in polyethylene bottles for subsequent determination. All tests were processed with duplicate and blank samples, and the analysis showed good accuracy for heavy metals, which was within ± 10%.

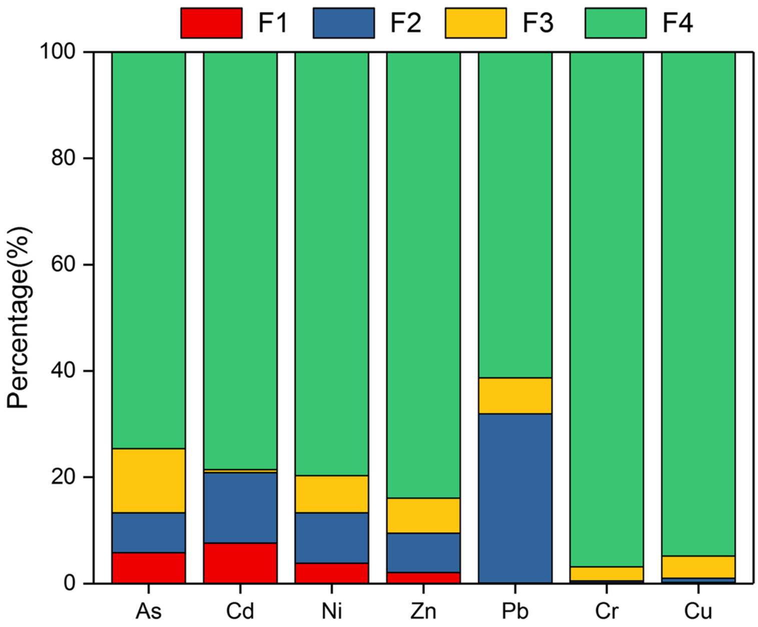
Risk assessment

Geoaccumulation index (I_{geo})

I_{geo} was a commonly used model to evaluate pollution level for heavy metals in soil since 1960s (Muller 1969). It was calculated using the follow equation:

$$I_{geo-n} = \log_2 \frac{C_n}{1.5B_n}$$

Fig 1 The chemical speciation of heavy metals in soil



where C_n represented the total content of heavy metal n (mg/kg), B_n was the geochemical background value for heavy metal n (mg/kg), and 1.5 was the coefficient to detect anthropogenic influence on the content of heavy metals in soil. The I_{geo-n} value can be classified for seven grades: $I_{geo-n} \leq 0$ no pollution, $0 < I_{geo-n} \leq 1$ no-to-moderate pollution, $1 < I_{geo-n} \leq 2$ moderate pollution, $2 < I_{geo-n} \leq 3$ moderate-to-heavy pollution, $3 < I_{geo-n} \leq 4$ heavy pollution, $4 < I_{geo-n} \leq 5$ heavy-to-extreme pollution, and $I_{geo-n} > 5$ extreme pollution (Muller 1969).

Risk assessment code (RAC)

As first proposed by Perin et al. in Perin et al. 1985, RAC had been widely used to evaluate the mobility and bioavailability of heavy metals in soil. RAC values were calculated as the percentage of the F1 + F2 fraction in soil. According to the calculated RAC values, risk could be classified to five levels:< 1% no risk, 1–10% low risk, 11–30% medium risk, 31–50% high risk, > 50% extremely high risk (Singh and Lee 2015).

Results and discussion

Soil characterization

The properties of collected soil samples were summarized in Table 1. It was observed that soil pH could affect the availability of heavy metals and microbial activity (Wadgaonkar et al. 2019). Soil samples in present study exhibited neutral pH with a mean value of 6.63, probably due to the release of acidic solutions during the coal mining process. Mean SOM value was 25.68 g/kg, which was higher than the average SOM content in the surface paddy soil of Anhui Province. Zhou et al. (2016) demonstrated that heavy metals could directly influence microorganism activities in soil thus inhibit the decomposition of organic matter, consequently leading to an increase in SOM. Based on the particle size analysis, percentages of clay, silt, and sand were found to be 7.35%, 32.06%, and 60.59%, respectively. Mean CEC value for soil was estimated to be 28.26 cmol/kg, while TN and TP content were measured as 3.7 and 75.63 mg/kg respectively.

Content of heavy metals in soil

Content of heavy metals in soil varied greatly for individual metal. As shown in Table 2, mean content of Cu, Zn, As, Cr, Cd, Pb, and Ni in soil was 64.39, 118.02, 23.42, 85.34, 0.54, 20.29, and 27.67 mg/kg, respectively. When in comparison with the background value of Huainan soil, heavy metals except Pb and Ni, were higher than corresponding background values. Furthermore, heavy metals in soil exhibited similar distribution characteristics with it in Zhangji coal (R^2 = 0.5998), which indicated that, to some extent, content of

Table 1 Characterization of soil properties

Index	pH	SOM (g/kg)	TN (g/kg)	TP (mg/kg)	CEC (cmol/kg)	Clay (%)	Silt (%)	Sand (%)
Mean value	6.63	25.68	3.70	75.63	28.62	7.35	32.06	60.59

heavy metals in soil was affected by coal mining activities. Besides, content of Cd ranged from 0.08 to 0.74 mg/kg with a mean value of 0.54 mg/kg, which was higher than that of the grade II environmental quality standard for soil in China and exceeded the average value of China, Anhui, and Huainan soil. The relatively high Cd content could mainly be attributed to the application of livestock manure and phosphate fertilizers in soil (Ouyang et al. 2017) (Table 3).

According to the I_{geo}, Cd and Cu exhibited moderate pollution, with mean values being 1.98 and 1.07, respectively. No pollution was observed for Cr, Pb, and Ni. The contamination levels of Zn and As were categorized as uncontaminated to moderately contaminated. Based on meta-analysis of data from 336 published articles, Huang et al. (2018) discovered that Cd from mining area was enriched in agricultural soil. These results indicated that Cd and Cu in soil were mainly affected by human activities instead of geochemical origin, which was also in agreement with previous study investigated by Zhou et al. (2007).

Chemical speciation of heavy metals in soil

Concentrations of the four fractions of heavy metals, which were indicative of metal availability and mobility, decreased in the order: acid-soluble (F1) > reducible (F2) > oxidizable (F3) > residual (F4). Chemical speciation of heavy metals in soil was depicted in Fig. 2 that revealed the potential risk of heavy metals in agricultural soil from mining area. The accumulative amount of four fractions was approximated to the total content of metals, which was in good agreement with our previous study on chicken manure that implemented using the same extraction scheme (Dai et al. 2018; Li et al. 2019). Generally, heavy metals investigated in present study were mainly in residual form. Besides, the proportions of acid soluble fraction were 7.85% for Cd, 5.84% for As, 3.86% for Ni, 2.05% for Zn, 0.20% for Cu, and 0.13% each for Pb and Cr, which indicated high mobilization for plant uptake once the environment conditions (e.g., pH, salinity, redox condition) changed. Some Cd could substitute for Ca ions in carbonates, and then bound with weak strength (Rosen and Chen 2014).

The reducible fraction of heavy metals ranged from 0.39 to 31.82%, with the highest value for Pb. This fraction could be released into the environment, especially when exposed to reducing condition. Therefore, the reducible fraction together with the acid soluble fraction had been identified as toxic and/or bioavailable fractions. The percentages of heavy metals in F3 and F4 fractions were in the range of 68.06–99.48%, which

Table 2 The contents of heavy metals in soil (mg/kg)

	Cu	Zn	As	Cr	Cd	Pb	Ni
Minimum	53.20	82.35	13.41	49.63	0.08	2.36	10.36
Maximum	87.64	132.60	35.62	94.34	0.75	30.20	42.91
Mean	64.39	118.02	23.42	85.34	0.54	20.29	27.67
CV%	32.5	25.6	12.1	24.3	19.5	42.8	16.4
Average background of Huainan soil[a]	30.69	58.35	–	91.53	0.06	23.52	32.03
Average background of Anhui soil	20.4	62.0	9	66.5	0.097	26.6	29.8
Average background of soils in China	22.6	74.2	11.2	61	0.097	26	26.9
Grade II environmental quality standard for soils in China[b]	100	250	30	200	0.3	300	50
Average content in Zhangji coalsa	14.78	25.87	9.71	41.63	0.07	12.89	13.7
I_{geo}	1.07	0.34	0.79	− 0.23	1.89	− 0.98	− 0.69

[a] Data from (Chen et al. 2011) and (Li et al. 2013); [b] Soil environmental quality standard (GB15618-1995).

Table 3 The content of heavy metals in soils in this study and other studies

	Cu	Zn	As	Cr	Cd	Pb	Ni
Mean	64.39	118.02	23.42	85.34	0.54	20.29	27.67
Huainan Xinji mining area soil[d]	23.08	52.57	–	57.23	–	20.37	–
National weighted mean content in agricultural soil[e]	28.4	83.4	10.8	62.4	0.24	32.0	28.4
Soil from China mining areas[f]	88.8	241.9	20.6	67.3	3.76	196.4	45.4
World soil[g]	30	90	7.2	40	0.35	2–300	20

[d] Data from (Li et al. 2008)

[e] Data from (Huang et al. 2018)

[f] Data from (Li et al. 2014b)

[g] Data from (Adriano 2001)

indicated immobilization and poor bioavailability (Nemati et al. 2011). Based on the result of F1 + F2, the environmental risk of heavy metals in soil was found to be in the order: Pb > Cd > Ni > As > Zn > Cu > Cr.

Numerous studies had reported that the result of chemical speciation of heavy metals could differentiate pollution sources. The F4 fraction had significant correlation with lithogenic inputs (Islam et al. 2015; Zhang et al. 2017). As shown in Fig. 2, Pb and As had lower percentage of residual fraction at 61.31% and 74.62%, respectively, indicating a higher rate of anthropogenic inputs such as industrial and urban activities. It was found that high Pb content in soil and sediment from Huainan City was due to the coal mining activities (Fang et al. 2015; Zhang et al. 2012). Besides, Xiong et al. (2015) highlighted that As levels in Huainan soil was mainly resulted from significant accumulation through long-term coal mining activities. Similarly, research on heavy metal content in soil near coal mining area in northeastern India also revealed that Cd and Pb were derived from coal mining activities (Reza et al. 2015).

Leaching characteristics of heavy metals in soil

Leaching of heavy metals in soil might cause groundwater contamination (Xiao et al. 2017a, b). The leaching capacity of heavy metals depended on several factors, such as soil type, nature of the contamination, and environmental conditions (Sahuquillo et al. 2003). Therein, chemical speciation of heavy metals was the primary factor influencing leaching behaviors (Wang et al. 2009). The results of the leaching experiment were shown in Fig. 4. Generally, the leaching tendencies greatly varied among heavy metals. Concentrations of Cu, Ni, Cd, Zn, and Cr in the leachates showed a decreasing tendency constantly with the increase in the accumulative simulated rain volume, especially for Cu, Ni, and Cd, with a sharp decrease in the first 105 mL. The maximum concentrations of Cu, Ni, Cd, Zn, and Cr in the leachates were found to be 4.56, 12.55, 0.26, 166.70, and 6.06 μg/L, respectively. Conversely, concentration of As and Pb increased with leaching time. During the leaching experiment, soil showed a lower pH value and reducing condition due to the input of NO_3^- and SO_4^{2-}, which resulted in the release of more heavy metals bound to Fe-Mn oxides into more mobile fractions (Shaheen et al. 2014, 2016; Xiao et al. 2017a, b). Results were similar to these reported in previous studies on dredged sediment and soil (Liu et al. 2018; Li et al. 2018). Compared to the quality standard for groundwater of grade V (GB/T 14848-2017), mean concentration of As in the leachate was higher than the maximum safety limit (50 μg/L), which indicated that leachate from long-term leaching may not be suitable for irrigation purpose.

In view of the leaching behaviors of Cu, Ni, Cr, Zn, and Cd, the whole process can be classified into two stages, wherein the initial stage showed a rapid decrease in elemental concentration in the leachate, followed by a slower rate of continuous decrease. The element leaching in the first stage was likely associated with the organic matter due to low adsorption energy in soil, which was easily released to the environment. The slower reaction may be due to the diffusion of heavy metal cations into soil micropores (Ma et al. 2010; Ma et al. 2006). Research on leaching behaviors of heavy metals from red paddy soil indicated that the exchangeable fraction correlated better with the leaching amount than acid-extractable fractions (Huang et al. 2015).

The leaching percentage of heavy metals under simulated rainfall condition was displayed in Fig. 3. In

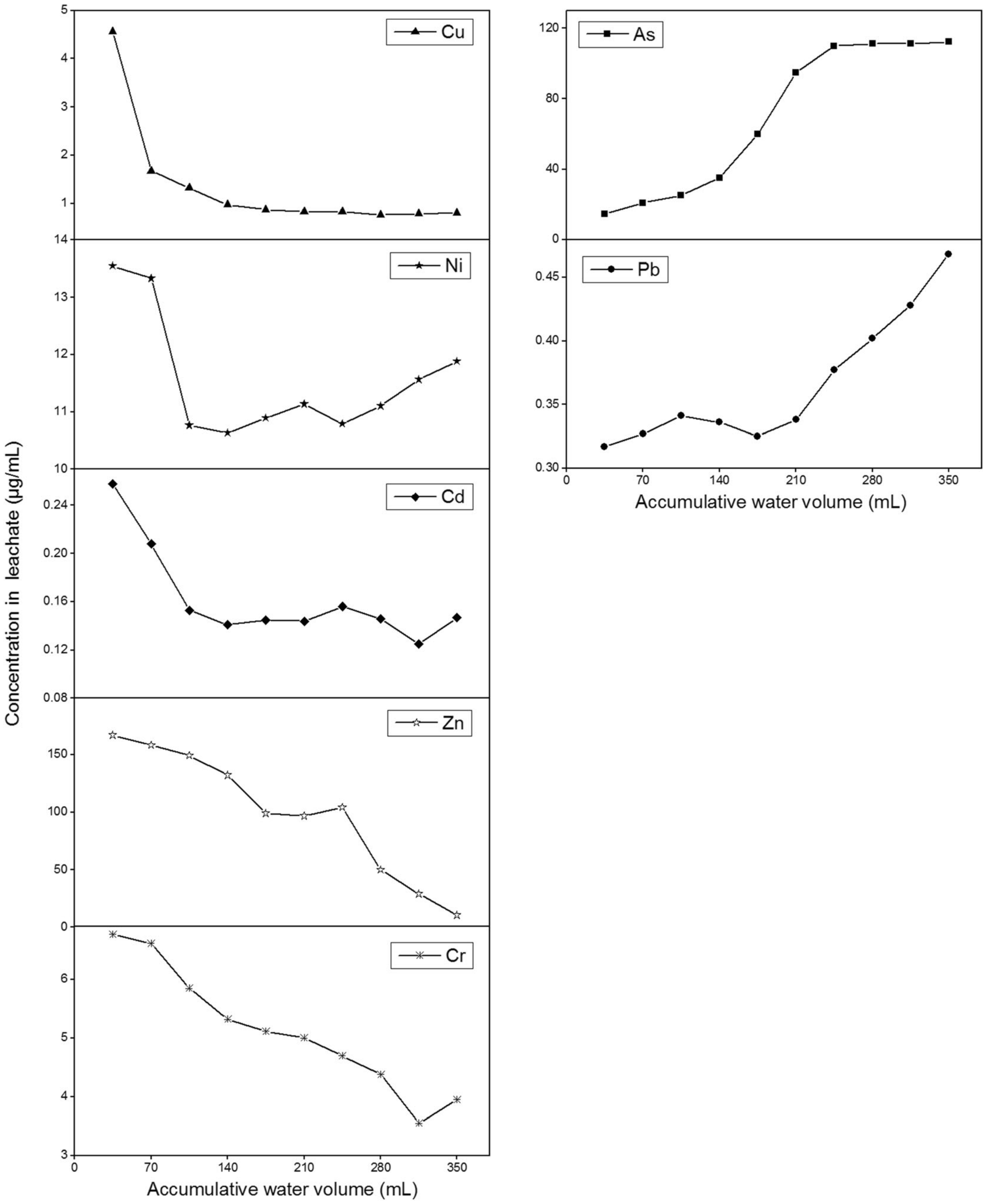


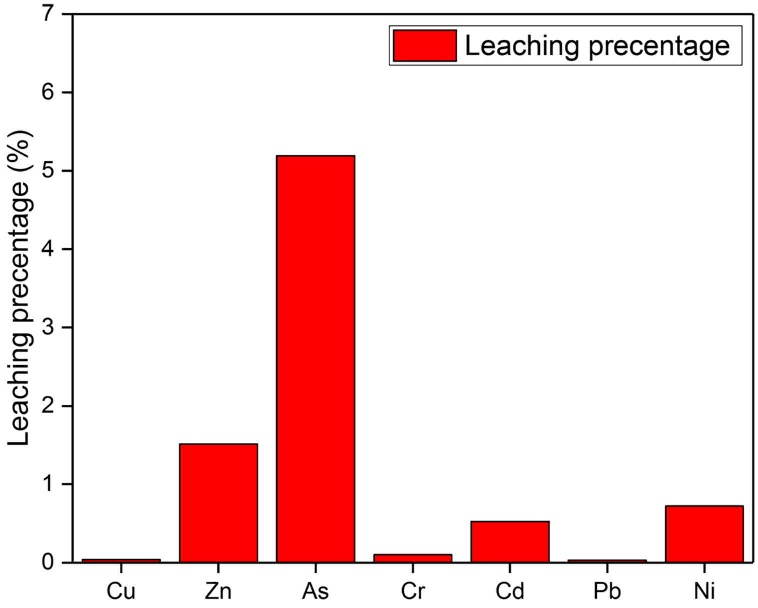
Fig 2 The concentration of heavy metals in leachates

reference to the total content of heavy metals in soil, the total leaching percentage ranged between 0.04 and 5.19%, which was mainly from soluble and easily exchangeable fractions (Li et al. 2015). It was found that As showed highest higher leaching ability thus posing higher environmental risk. Therefore, more attention should be paid to As, as it might cause contamination in deeper soil layer and watershed of paddy field. Apart from As, the leaching percentage of rest heavy metals followed the sequence: Zn > Ni > Cd > Cr > Cu > Pb.

Environmental risk assessment of heavy metals in soil

Environmental risk assessment of heavy metals in soil was implemented using the risk assessment code (RAC) and the result was illustrated in Fig. 4. It was notable that

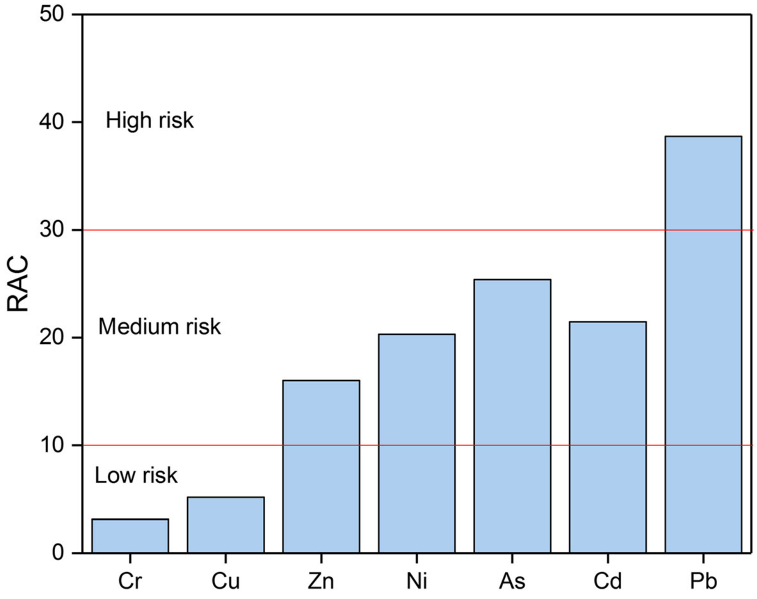
Fig 3 The leaching percentage of heavy metals in soil under simulated rainfall



all metals pose potential risk (RACs > 1%) in soil. Lead was found to pose the highest risk due to its high reducible fraction, followed by Zn, Ni, As, and Cd, which posed medium risk, although the content of Cd exceeded the environmental quality standard for soil in China. Furthermore, Cr and Cu were found to pose low risk.

According to the study of Jiang et al. (2014), soil near coal gangue dumps also had more severe Cd pollution, and exhibited high potential ecological risk. Shang et al. (2016) also indicated that Cd had the highest ecological risk due to the effect of coal gangue and fly ash. In the present study, the paddy field was far away from the coal gangue pile and fly ash, thus those aforementioned influencing factors were excluded. Nevertheless, heavy metals, especially Pb, As, Cd, Zn, and Ni in soil would pose high environmental risk, which may easily enter the food chain that deserved further attention.

Fig 4 The RAC for heavy metals in soils



Conclusions

Characteristics of seven heavy metals in soil samples collected from Huainan coal mining area were analyzed. Mean content of heavy metals followed the sequence of Zn > Cr > Cu > Ni > As > Pb > Cd, wherein Cd exceeded the maximum value of grade II environmental quality standard for soil in China. The results of chemical speciation indicated that most of heavy metals were retained in the F4 fraction; whereas Pb, As, and Cd had lower residual fraction content, which indicated that these heavy metals in soil was impacted by coal mining activities, i.e., from anthropogenic sources. According to the result of risk assessment, studied heavy metals would pose potential risk to the environment, among which Pb posed the highest risk. Besides, the simulated rainfall leaching experiment revealed that As was the most leachable element, with a leaching percentage of 5.19% and may result in underground water contamination under long-term leaching.

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